French Atomic Energy Commission
Nuclear Energy Division
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Testimony of Jacques Bouchard
given before the Committee on Energy and Natural Resources
on July 18, 2001

Mr Chairman and members of the Committee,

Thank you for this opportunity to testify on the important issue of reprocessing for the future of nuclear energy.

There are few doubts that nuclear fission will still play a role in the satisfaction of future energy needs around the world. It is among the solutions which are proposed in the recent NEPD report and it has been mentioned by both your President and your Vice President. It is also an important point of the European Union green report issued last November and it is part of the future plans for energy in Asian countries, in particular Japan and China.

The results obtained with the 400 existing nuclear reactors show that nuclear electricity production is economically competitive and with a very low impact on the environment. The safety records for the last fifteen years are most satisfactory and should allow the public to forget the lack of experience which led to Three Miles Island and the failure of society which made possible the Tchernobyl disaster.

The only problem which remains a real difficulty in some countries, France in particular, is the management of highly radioactive wastes. For most of the people, the actual concern is the long term behaviour of long lived radioactive elements. In this respect, reprocessing of spent fuels is a key point as it allows a strong reduction of both the volume and the long term radio toxicity of wastes.

Historically, reprocessing of spent fuels, followed by recycling of valuable materials, uranium and plutonium, was intended to increase the use of natural resources. It was part of a scheme which included breeders in order to extract most of the energy contained in natural uranium. That remains a clear objective for the future. With existing light water reactors, we burn only 1% of the natural uranium and we let aside 99%, either in provisional storages or in waste disposal. If we don't improve the situation, with increasing energy needs, we shall exhaust in a few decades the uranium resources, at least those which can be recovered at a reasonable price. Thus, reprocessing is a cornerstone for satisfying future energy needs.

In shorter terms, it is also a key point for the waste management. In any other activity, a good waste management policy includes selection of various types of wastes, recycling

of what can be reused and disposal solutions adapted to each kind of product. Nuclear wastes should not be an exception to this basic rule.

For France, nuclear spent fuel is not even a waste as it still contains a huge amount of energy valuable products. The way we treat it at the output of the reactor is of major importance for the waste management policy. With present technologies, the fuel unloaded from reactors still contains 95% of uranium and 1% of plutonium. It contains also 4% of actual wastes, fission products and minor actinides.

Reprocessing allows us to separate uranium and plutonium from the actual wastes and then, with the vitrification process, to put these wastes in a robust containment for long term storage or final disposal. It is basically a wise policy for waste management.

The main argument often opposed to this policy has been that by extracting the plutonium we could open various possibilities of diversion and thus we may create a weakness in the non proliferation policy. Let me try to bring some consideration in this discussion:

- First, we are certainly not underestimating the risk of proliferation and we are fully sustaining the various measures which are taken on an international basis to try to avoid this risk.
- The plutonium coming from light water reactors, a large majority of existing production facilities, is not at all well suitable for nuclear weapons but we agree on the fact that we cannot completely exclude a wrong use of it, even if it will be much more difficult than other proliferation routes.
- A reasonable way to limit the risk, while taking benefit of reprocessing, is to burn the plutonium as soon as possible after extracting it from spent fuels. It is what we are doing in France. The plutonium extracted at La Hague is used to fabricate MOX fuels and we have presently 20 reactors loaded partially with MOX fuel. That means that, except the necessary hold-up for recycle management, we have no plutonium on shelves.
- Therefore, the diversion risk is limited to the operations themselves, output of the reprocessing plant, transportation and MOX fuel fabrication. There, we have very strict domestic and international controls and we are fully convinced that they are suitable to avoid any significant diversion.
- Last but not least, we consider that from the non proliferation point of view it is better to burn plutonium rather than to keep it in store, even if it will be quite difficult to recover it from stored spent fuel with existing technologies. In other words, we think it would not be easy to explain to French people that we should have to dispose of hundreds or thousands tons of plutonium underground somewhere in the country.

Now, looking to the future, assuming that nuclear energy will still be needed, very probably on a larger scale than presently, reprocessing will more than ever be necessary for both economy of resources and waste management. The existing technology of which we have now a large industrial experience, has proved to be efficient and economic. But progress should be made and we are working on it in the same way as we are working on future reactor designs.

Taking into account the present concern on waste management, while assuming the problem of plutonium is completely solved by reprocessing and recycling, we should consider the possibility of destroying the other actinides, the so-called minor ones, neptunium, americium… Several countries have important R&D programs on partitioning and transmutation. We have already succeeded in developing complementary processes which could be implemented in reprocessing plants to extract those minor actinides. We know how to burn them, either in reactors or in accelerator driven systems. For the future we should try to develop an integrated approach based on recycling of all the actinides in such a way that the actual wastes to be definitely disposed will only be the unavoidable fission products, the amount of which is directly related to the energy production.

Another improvement will be to limit as much as possible transportation of radioactive materials. An objective could be to have reprocessing and fuel fabrication on the same site and not too far from the reactors.

Technical solutions can be developed, either improvements of existing technologies or developments of new ones such as, for instance, dry processing or pyroprocessing which has been successfully tested in your country in the frame of the Integral Fast Reactor studies developed by Argonne National Laboratory.

Mr Chairman, as a conclusion, I would say that reprocessing will be sooner or later a necessity for use of nuclear energy in a sustainable development. It’s already an efficient tool for waste management and in some countries an industrial reality. It can certainly be improved to be still more efficient, more proliferation resistant and cheaper.

Thank you for your attention.

A N N E X

REPROCESSING OF SPENT FUELS :

STATUS OF FRENCH EXPERIENCE AND CURRENT DEVELOPMENTS.

INTRODUCTION

Spent fuel released of the French 58 PWR plants (1200T/year) still have a major energy potential that is greater than the equivalent of 24 million tonnes of oil/annum in France, when used in light water reactors (and more than 50 times greater in fast breeder reactors).

On the other hand, spent fuel has a long term potential radiotoxicity (more than 100 000 years to fall to the level of natural uranium), whose main components, in decreasing order of importance, are plutonium, the minor actinides and the fission products.

Nuclear energy is able to provide a durable solution for the very high stakes involved in humankind long term energy supply and sustainable development : long term resources (several thousand of years, if the fuel use is optimised by spent fuel processing and uranium-plutonium recycling) and non greenhouse effect or toxic gas emission. On the other hand, many in the public opinion consider that the major problem currently posed by nuclear energy is that of the safe long-term management of radioactive waste.

We consider that **reprocessing and recycling** is both an **attractive and responsible solution for the** nuclear energy **fuel cycle**, for the main following reasons :

- it allows **major energy resources saving** and optimisation,
- it makes it possible to **drastically minimise** the **quantity and** the long term **radiotixicity of** High Level and Long Life **Waste**, and opens several genuine ways of improvement in this field,
- it is compatible with a **proliferation resistant** approach,
- it is based on a safe and **at maturity industry**, which takes benefit from a very satisfactory experience feedback, continuous optimisation and high level R&D.

I - SPENT FUEL MANAGEMENT

Characteristics of spent fuel

Spent fuel based on uranium oxide released after three years of irradiation in one of the EDF pressurised water reactor plants contains (figures provided for a combustion rate of 47.5 GWj/t):

- Heavy nuclei having energy value, whether they are fissile (U_{235}, Pu_{239} and Pu_{241}) or fertile (U_{238}) and representing 96% of the heavy nuclei irradiation balance,
- Elements that have undergone fission (products of fission) and elements that have undergone degradation through neutron capture (pair isotopes of uranium and of plutonium, minor Am, Cm and Np actinide series) that make up 4 % of this balance.

To this has to be added the activated and slightly contaminated technological waste made up of fuel assembly sheaths and fittings.
The main components of long-term toxicity, in decreasing order of importance, are plutonium, the minor actinide series and a few long-lived products of fission.

Management options

After the spent fuel has been allowed to cool as it stands to a level where it can be managed in the long term, there are two major options that can be considered for the downstream cycle of nuclear fuels:

- the open-ended cycle where spent fuel is regarded as global waste. This option consists in packaging it in sealed containers before placing it in direct storage in a deep geological repository;
- the closed cycle that includes spent fuel reprocessing, the recycling of valuable materials and the specific packaging of elements deemed to constitute waste at the end of a selective sort process. These packaged waste are the residues generated by these operations and are destined for geological disposal.

Interim storage of spent fuel is made possible by the robust nature of water reactor fuels and by suitable containers. This allows decision-making processes linked to these two options to be « time-flexible ».
The grounds on which each country chooses one of these options vary and are frequently tied in to the issue of plutonium management. The reprocessing – recycling option is part of long-term energy strategy, of an approach aimed at reducing any potential nuisance caused by ultimate waste, or by a reduction in stocks of fissile materials from military sources. The direct disposal option often goes with a nuclear withdrawal policy or with the wish to avoid isolated plutonium through fear of proliferation.

II - THE ADVANTAGES OF THE REPROCESSING POLICY

Waste management

In the long term (beyond 200 years), plutonium contributes to approximately 90% of the radiotoxic content of spent fuel fuels. Reprocessing the fuel make it possible to separate the valuable materials (uranium and plutonium) from the final waste, in which the remaining quantity of plutonium is limited to an absolute minimum (typically less than 0.1% at the end of the La Hague plant's process). (Fig. 1).
The ultimate radionuclides in the waste (fission products and minor actinides) are vitrified [1](integration of these radionuclides in a vitreous matrix), in order to obtain a small volume of waste packages (~150 m3/year of vitrified waste in France) offering very high performance and long lasting confinement characteristics : R&D work has proven that water action on the glass would be negligible even after several thousands of years.

[1] 99% of the radioactivity of the radionuclides in the final waste are within the vitrified wasteforms ; about 1% is conditioned in compacted wasteforms (structure waste [hulls and end-pieces] and technological waste)

Saving natural resources

Fissile nuclei obtained from the reprocessing of spent fuels have a major energy potential that is greater than the equivalent of 24 million tonnes of oil/annum in France.

Plutonium is recycled in the form of a combined fuel containing uranium oxide and plutonium oxide (Mox). Current policy in France consists in recycling plutonium in the CPY 900 MW reactors combined with adequation between the amounts of separated plutonium and the amounts re-introduced into the reactor[2]. This balance in plutonium throughputs results in the annual reprocessing of 850 tonnes of spent fuel of the 1200 tonnes produced by the French nuclear electricity generating plants. The remainder is stored at the power stations pending subsequent reprocessing.

The production of energy from plutonium, even if it is less efficient in a water reactor than in a fast neutron reactor, constitutes by itself a full justification for the recycling of plutonium in current reactors. This plutonium recycling process allows us to generate approximately 30 TWhe/an or approximately 7% of annual output (≈ 400 TWhe/annum), thus reducing our requirement for natural uranium (a saving of approximately 5% in the case of a plant that has a 45 year life span). This first mono-recycling also enables us to reduce, by approximately 20%, the quantity of plutonium produced each year, and constitute the first step towards plutonium inventory stabilisation (fig. 4).

The uranium discharged from spent UO_2 fuels is processed at Pierrelatte and at Romans and stored. Its properties are similar to those of the ore and, therefore, it can constitute a fuel resource. In this case, it still requires enrichment to compensate for the presence of isotopes other than uranium 235 and 238. EDF has been recycling some reprocessed uranium since 1995 in the Cruas 3 and 4 reactors that are fully charged with fuel containing recycled uranium.

Optimised use of storage facilities

Mono-recycling of plutonium allows us to concentrate the plutonium radioactivity in the spent Mox fuels in a small volume (a factor of 7 compared to UO_2) : one Mox assembly is made with the plutonium obtained from the reprocessing of seven UO_2 assemblies. This policy has the advantage of using existing storage facilities (fuel buildings for the reactors, water pools at La Hague) where, on the basis of current throughputs, there will be no new requirements before 2015.

III - PRESENT STATUS OF REPROCESSING IN FRANCE

Industrial maturity

The French reprocessing tool has been built and has shown a very satisfactory feedback of experience. This is one of the industrial areas where the technological progress achieved by France is particularly acknowledged. Let us for instance note that more than 18 000 metric tons of LWR spent fuel have been reprocessed at the La Hague plant

Experience acquired in reprocessing and recycling have enriched the technical knowledge of the nuclear industry world-wide and led to the optimisation of processes used, to enhanced performance and reliability of control systems, to increased control over operational safety as well as safety in maintenance and operation in active zones. A series of major improvements has been made to reprocessing operations, resulting in the optimisation of the amounts of waste (B waste has been reduced by a factor of 3 since 1991), and a drastic reduction in discharge (activity discharged in liquid and gas waste from the La Hague site reduced by a factor of 10 over a period of 10 years) (fig. 2 and 3). These efforts are being pursued with the development of complementary operations in preparation for the entry into force in 2020 of the obligations imposed by the Ospar convention. Further, the reprocessing system used (Purex) constitutes

[2] In effect, we have to take into account the phenomenon consisting in the conversion of Pu 241 into Am 241 which is a neutron poison and gamma emitter. This means that the amount of separated Pu awaiting recycling has to be restricted to approximately 2 to 3 years of consumption and that we have to optimise the on line procedure between the separation of Pu, its availability for producing Mox and delivery of new Mox assemblies to the NPP for loading into the reactor.

the first step in an advanced process for the separation of the main long life radionuclides studied as part of line of work 1 of the law of the 30 December 1991 (Appendix 2).
It should also be noticed that feedback of experience has shown the good behaviour of Mox fuel in the reactors' cores and the absence of any impact on plant operations and environment .
Spent Mox fuels are also adequate for reprocessing, using the present process implemented by La Hague (11 t of Mox have already been processed using this technology).

International expertise

The reprocessing technology, industrial operation and policy is not limited to France. Japan, who has developed the technology in the seventies with the Tokai fast reactor spent fuel reprocessing facility, and is now building the reprocessing plant at the Rokkasho-Mura site, using COGEMA/CEA technologies. In the UK, UKEA has developed the Dounreay facility for reprocessing the spent fuel from the fast reactors DFR and PFR. The BNFL THORP reprocessing plant started operating in 1994, and since reprocesses irradiated oxide nuclear fuel from Japan, British, and other European water and gas cooled reactors.
The USA and Russia have clearly indicated their interest in recycling technologies, for the consumption in civilian reactors of significant amounts of plutonium from surplus nuclear weapons: "burning" military plutonium in reactors means that, in effect, it reverts to the properties of civil plutonium and thus becomes inappropriate for re-use in a military context, while generating electricity.

Economy

The conclusions of the latest OECD figures show that, at this moment in time, downstream cycle considerations contribute the following to the cost per nuclear kWh : 0.21 cts with reprocessing and 0.13 cts for direct storage of spent fuels. In addition, the investment has been made in the La Hague plant and it is operating satisfactorily. Similarly, overall future technical progress on fuel, cycle processes (especially spent fuel reprocessing) and on reactors, arising from R&D initiatives, must be taken into consideration when carrying out an economic evaluation.
The Charpin-Dessus-Pellat last year's report has similarly emphasised the modest economic impact made by downstream and end of cycle considerations on the cost per kWh (0.23 cts on the price of a kWh which is approximately 1.95 cts).
This all goes to show that the financial penalty created by spent fuel reprocessing compared with a potential direct storage remains extremely low (less than 0.13 cts per kWh) in the face of what is at stake environmentally and of the impact of fluctuations in fossil energy prices (gas and oil) on the cost per kWh.

Proliferation resistance

Compared to one open cycle, reprocessing and recycling avoid to dispose in the French subsoil 100 000 metric tons of spent fuel containing 1000 metric tons of plutonium (figures corresponding to one century of nuclear energy production in France), which would otherwise represent a real "plutonium mine" lasting in the very long term.
At the opposite, reprocessing spent fuel and multirecycling the plutonium both consume it and makes its isotopics quite inadequate for weapon purposes. Let us note that improvement could be considered in the fuel reprocessing industrial technology in order to absolutely avoid any isolated plutonium : it can consists in a simultaneous conversion (co-precipitation) of liquid mixed uranium and plutonium into intimate uranium – plutonium oxide powder, the directly transformed into MOX pellets.

Furthermore, all the steps of the fuel cycle (enrichment, fuel fabrication, at reactor operations, transportations, reprocessing and back end, …) are under national and international safeguards. The corresponding controls are strict and allows us to be confident in the absence of significant diversion.

IV- CURRENT STRATEGY FOR DEVELOPMENT

Increased performance for UO_2 fuels

UO_2 fuels used at present have average combustion rates of approximately 45 GWd/tU. Ongoing progress made in fuel design, feedback on experience and development initiatives have resulted in increased safety margins. These have made qualification data available on advanced fuels and allow us to anticipate a gradual rise in burnup to approximately 57 GWj/tU, on average, during the next decade. The increase in average burnup will lead the gradual reduction in the amount of irradiated UO_2 fuels. In time (by 2015 present forecast), and on the hypothesis of the amounts of spent fuels currently processed (850 tML/year[3] of processed UO_2 for approximately 100 tML/year irradiated Mox), this will allow us to achieve a balance in flow between discharged and reprocessed UO_2 fuels while keeping the amount of Mox at current levels.

Improved Mox fuel performance

Optimised fuel management and the use of nuclear reactors will result in the development of a new Mox fuels management system said to be "on par" with that of the UO_2 fuel. The object of this "parity" is to achieve an energy and economic balance between Mox and UO_2 assemblies. To achieve this, the Mox assemblies must reach energy levels equivalent to those of current UO_2 assemblies that are 3.7% enriched and thus allow for average irradiation of approximately 45 GWj/tML. This will result in a Mox fuel having a maximum plutonium content of 8.65%. For an annual quantity of approximately 100 tML/year of Mox, this would mean that we could easily maintain a balance between separated and recycled plutonium throughputs, while producing approximately 10% of nuclear power output (approximately 40 TWhe).
This enhanced performance of the Mox fuel plays an essential role in ensuring that recycling is a cost effective process and in obtaining improved levels of plutonium consumption by reactors (reduced amount of plutonium residue per TWhe).

Multiple recycling and stabilisation of the plutonium inventory

Safeguarding natural resources and minimising the harmful nature of ultimate waste products should allow us to confirm the validity of pursuing the line of sustainable development of nuclear energy. However, complete control over the plutonium inventory, that is to say, a balance between the throughput of plutonium produced and consumed each year, cannot be achieved only by the Mox mono-recycling option. Consequently, this stabilisation of the inventory requires the implementation of complementary technical solutions that perform better again than Mox.
The French nuclear plants are relatively young (on average, 15 years old). At present, reactors have an average life expectancy of 40 years. However, this limit can be extended to 45 and possibly to 60 years which would mean that these facilities would reach the end of their lifetime in about 2050. In addition, the next reactor we consider to be built is still a light water reactor, the European Pressurized Reactor, whose average life expectancy is 60 years. Therefore, we have to focus some research effort into the possibility of achieving plutonium inventory stabilisation in water reactors alone. There are a number of existing solutions that only affect the fuel and its cycle but that require a validation programme spread over several years.
A first solution is derived from current plutonium fuel technologies. It consists in new plutonium assemblies, called CORAIL, with islands of standard UO2 rods surrounded by Mox rods. Such fuel could for instance be initially implemented in the existing reactors before 2020. A second solution requiring further research and development into the fuel could conceivably be implemented in about 2030. It consists in advanced assemblies, called APA[4], an heterogeneous bundle including UO2 rods and annular rods made of plutonium oxide within an inert matrix. In addition to controlling the plutonium inventory, it would reduce its overall quantity in the fuel cycle and, potentially, allow us to limit the inventory of the minor actinide.

[3] tML : tonne of heavy metal
[4] APA Advanced Plutonium Assembly

Advanced partitioning and transmutation

Current research and development work in hand as part of the law of the 30th December 1991 on waste, which results are due in 2006, specifically addresses partitioning, transmutation and conditioning of radionuclides and open the way for even more specific management routes by radionuclides.

Studies have led to the definition of an advanced process for separating the minor actinide (fig. 5). After plutonium, these elements have the highest radiotoxicity levels. The fact that a spent fuel reprocessing industry already exists makes it realistically possible that, from 2015, we could see the application of this process for producing vitrification packages that could only contain fission products whose potential radiotoxicity would then decrease to the initial natural uranium level after a few hundred years (instead of approximately ten thousand years). Thus, freed of heat generating actinides, after a typical 200 year storage period, the vitrified packages would reach very low thermal output levels so that they could be disposed in a geological repository, with a quite low radiotoxic inventory, and without the need for any specific heat limitations.Minor actinide series could be stored in safe conditions for the time needed to develop appropriate management methods (transmutation or specific conditioning).

Transmutation studies are examining the conversion of long lived radioactive atoms into non-radioactive atoms or radioactive atoms with a much shorter life. They have highlighted the possibility, considering different reactor types (light water reactors, fast spectrum reactors and/or transmutation dedicated systems), to control plutonium and minor actinide inventories over timescales of twenty to fifty years, and drastically reduce the long term radiotoxicity of final waste.

Studies into the packaging of ultimate waste to improve its long-term containment, in addition to the industrial vitrification of HLW, has demonstrated the value of new matrices specific to separated radionuclides.

Thus, technical and scientific developments arising out of research work carried out as part of the law of 1991 have led to additional methods for managing long-lived HLW. All these new methods involve achieving and maintaining control over the spent fuel reprocessing industry. Their industrial application is only obvious as the continuance of the La Hague plants. In the meantime, continuing to process spent fuel at La Hague enables us to only produce plutonium free waste, packaged in small containers, guaranteeing a high performance and durable containment (vitrification for the most radioactive and long lived radioactive elements).

CONCLUSIONS

In conclusion, the legitimacy of using nuclear power as part of sustainable development in order to meet future energy needs, rests on our ability to demonstrate that we are capable of safely managing nuclear materials and especially spent fuel and waste. Whatever the path we choose, direct storage or treatment-recycling, the closure of the nuclear cycle will require us to create a geological waste repository. Public opinion will then be entitled to expect that we have used our best endeavours to minimise the long term residual effect and this, all things being equal, means that we have to limit the quantity and radiotoxicity of waste stored in those depositories.

On this issue, the reprocessing-recycling approach presents conclusive advantages over the direct storage of spent fuels. Multiple recycling of plutonium allows us to diminish by several units (factor of 3 to 10 depending on cooling time) long-term radiotoxicity of long-lived elements; savings could even be greater, by several magnitudes, if we include the additional recycling of minor actinide series. On the other hand, in the direct storage option, spent fuels that have not been designed to form the ultimate packaging for long-lived radionuclides, will retain, in the long term, an energy potential that remains unused and a very significant radiotoxic inventory.

The reprocessing of spent fuels and their potential developments form the keystone of a strategy that has set its sights, at the very least, on achieving a control over the plutonium inventory in the fuel cycle, motivated by the drive to provide our environment with the maximum possible protection and to save on our use of energy materials. Even now, we can foresee solutions involving the recycling of plutonium in water reactors other than Mox, alongside a modest financial impact[5]. By using these solutions, we could gradually increase the amount of plutonium burned until we stabilise its inventory at the end of a twenty to thirty year period.

[5] The financial impact, in the region of 0.13 cts per kWh, do not compare with production overcosts currently accepted in respect of renewable energy sources (6.5 cts / kWh wind generated power in France or 1 DM (44 cts) per kWh photovoltaic power in Germany compared with the costs per kWh nuclear power that would increase from 1.95 cts to 2.07 cts).

Fig. 1 Balance of processing operations in the PUREX process

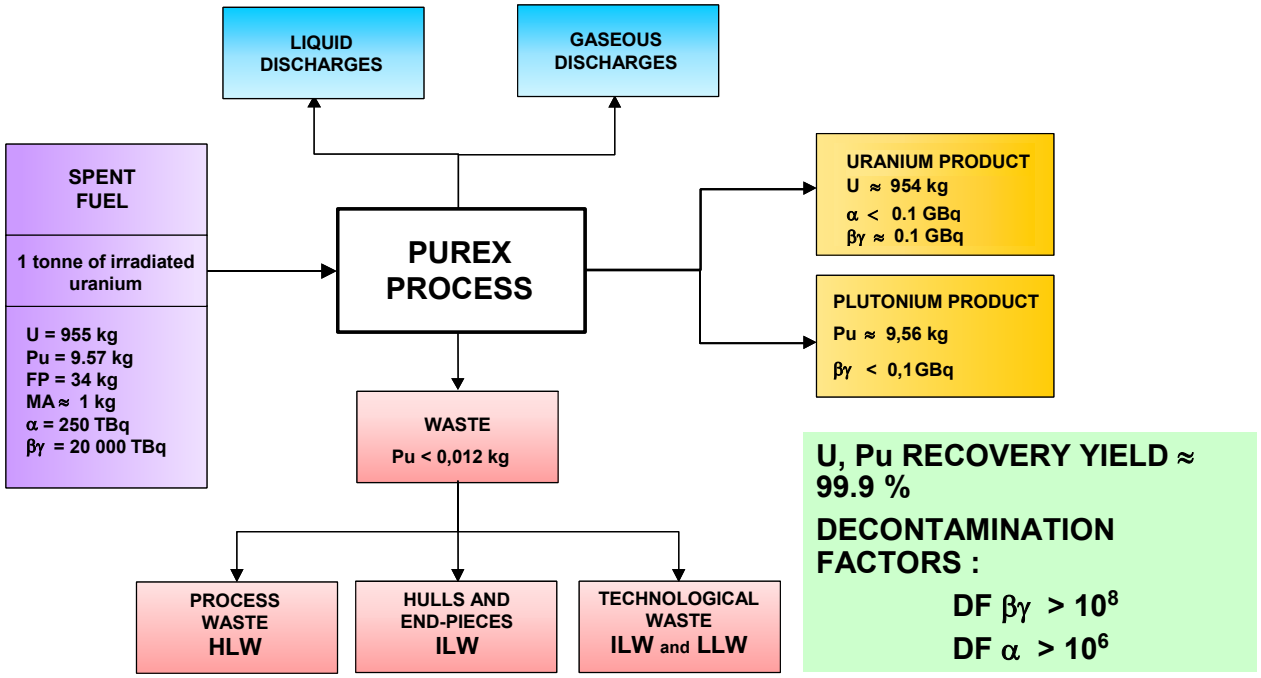
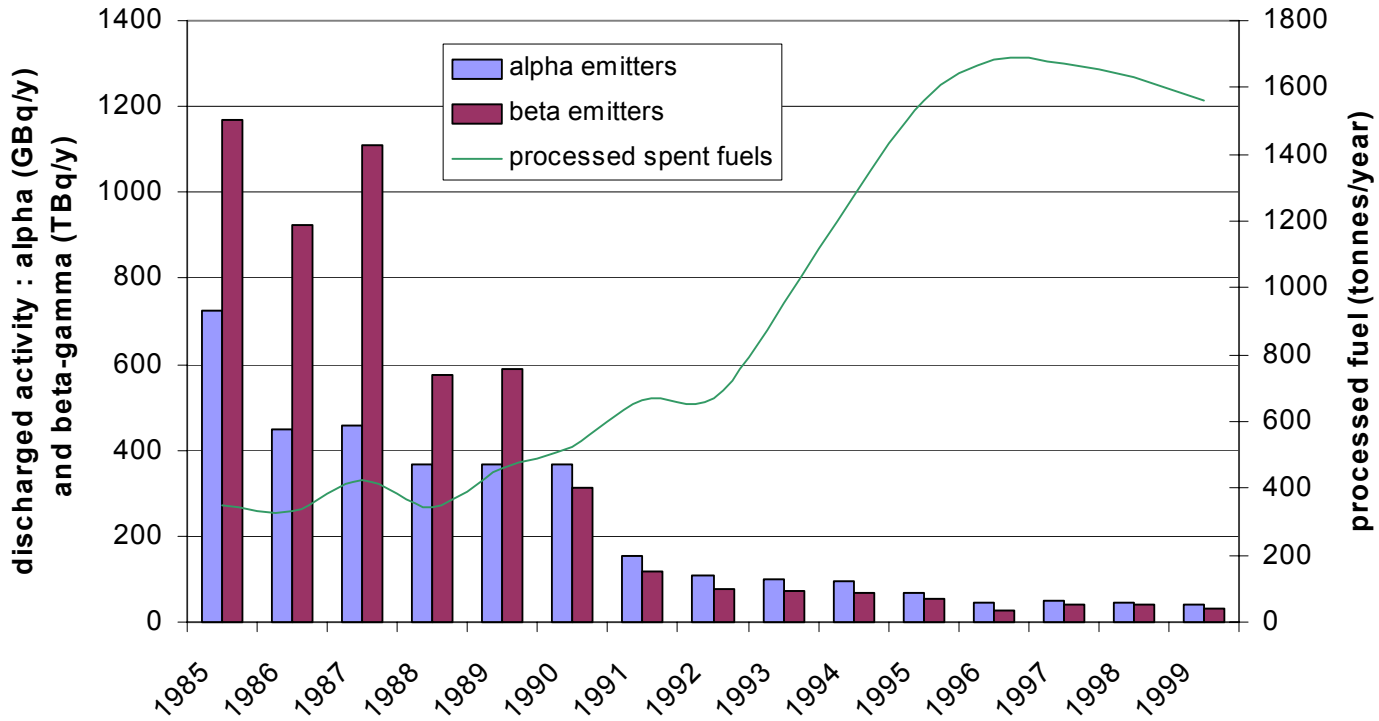


Fig. 2 Liquid discharges from La Hague site



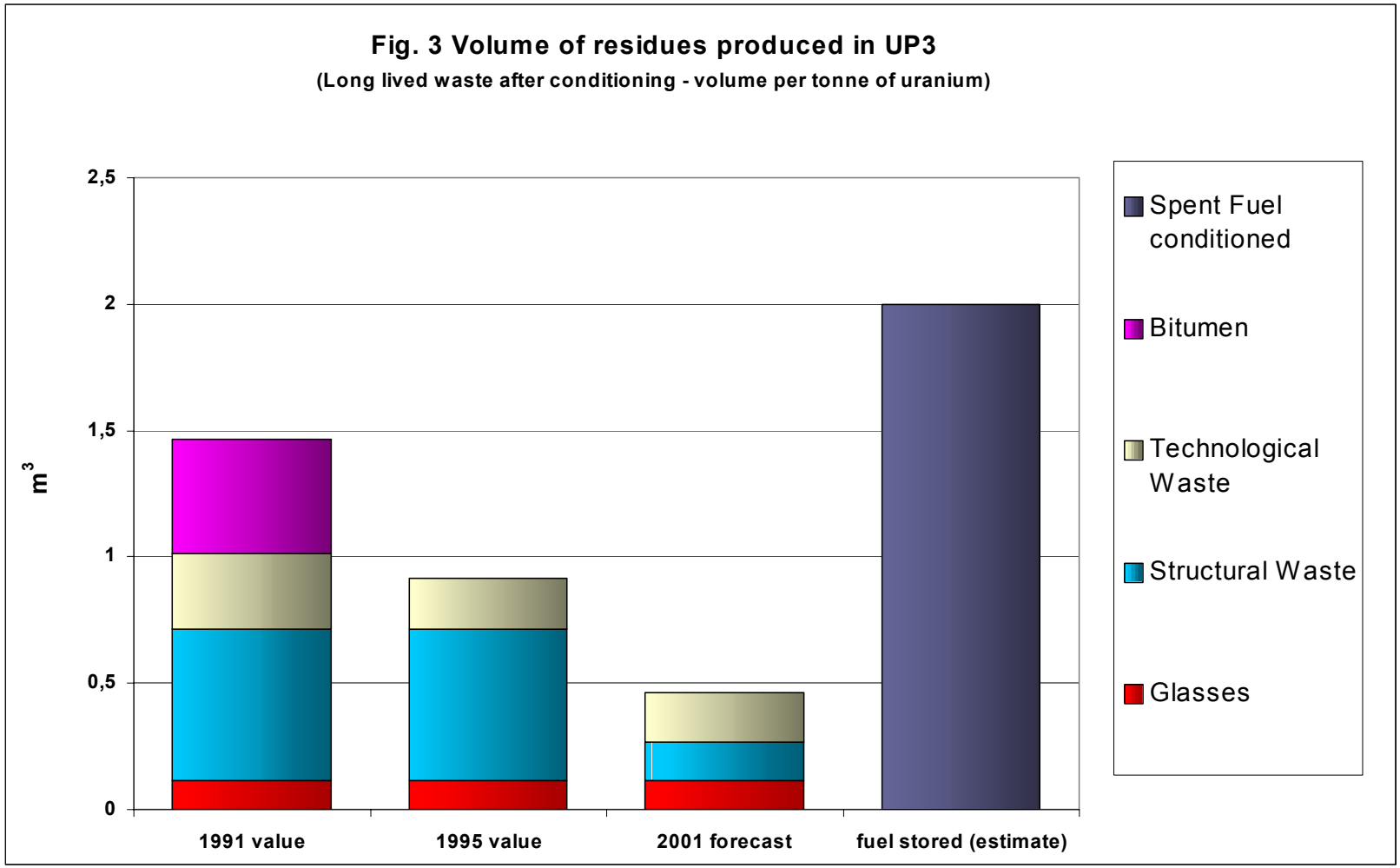
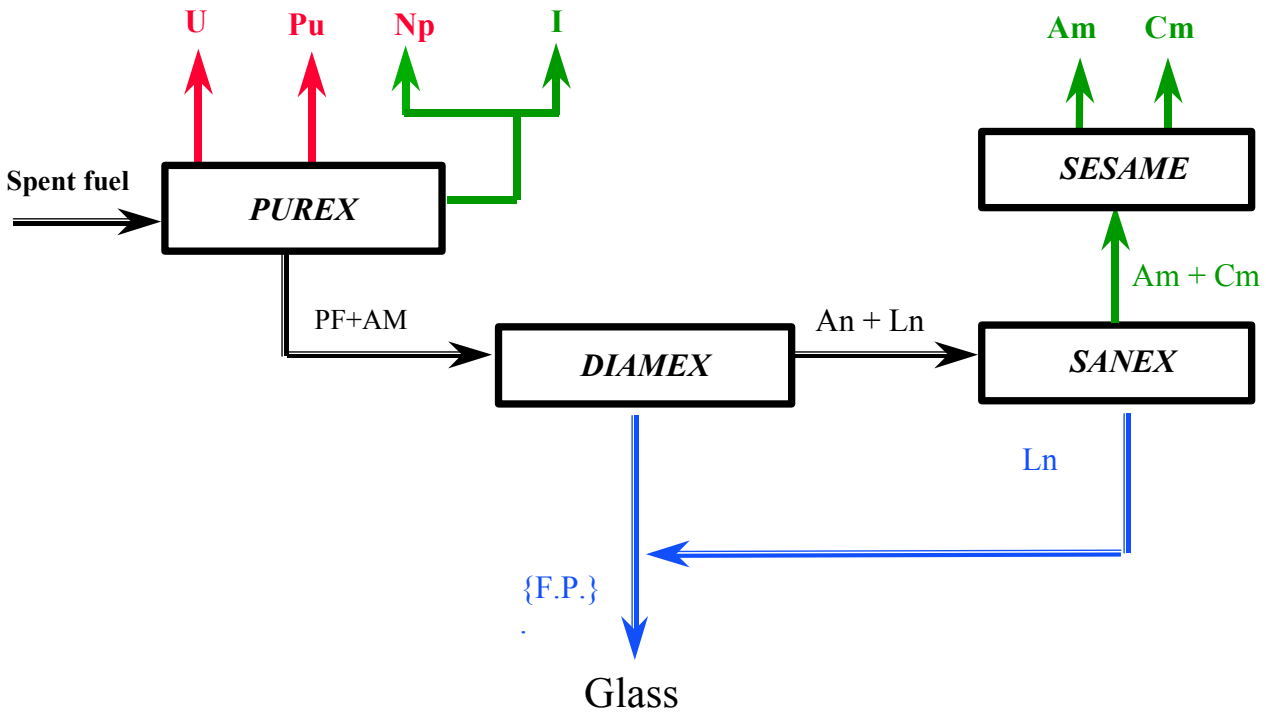


Fig.4 : Plutonium recycling in PWRs (400 TWh/year)

Fig. 5 Advanced partitioning scheme



Appendix1 : Industrial development of the spent fuel reprocessing technology

1.1 General remarks on reprocessing methods

The reprocessing methods were originally developed to produce plutonium for military use.

The first process used on a large scale was to entrain the plutonium by a precipitate of bismuth phosphate. It was used in the United States as early as 1945. This discontinuous operation had to be repeated several times to attain the required purity.
The use of solvent extraction as a separation method was a decisive advance, because it allowed continuous counter-flow operation on homogeneous phases, achieving very high separation factors.

Various solvents have been used, but the PUREX process (for Plutonium Uranium Refining by Extraction) quickly supplanted all others and is used in all existing plants. In particular, it is the only process ever used industrially in France, from the first UP1 plant at Marcoule, started up in 1958, to the UP3/UP2-800 industrial complex currently in operation at La Hague.

The success of the PUREX process has not prevented parallel study of other processes, in particular processes using a non-aqueous route, aimed at reducing the number of operations and the volume of the installations by eliminating water and organic compounds through the use of a mineral reagent that is stable when exposed to radiation.

The most important developments have concerned:

- Fluoride evaporation processes using the differences in vapor pressures of the fluorides of the elements present in the spent fuels. These processes are no longer being investigated.

- Pyrochemical processes using molten salts and metals: they have the advantage of high density of the installations, because of the possibility of working with high concentrations of nuclear materials and the small number of operations. But their implementation entails many technological difficulties arising from the use of very aggressive compounds (chlorides or fluorides) at high temperatures. Furthermore, the performance levels of these processes are still modest - substantially inferior to that of the PUREX process.

They are still the object of major research efforts in France, the USA, Japan and Russia.

Brief description of the PUREX process

The PUREX process is based on the selective extraction of uranium and plutonium by an organic compound, tributyl phosphate or TBP. It today benefits from more than a half century of research and development since the concept was established in the 1940s in the United States.

The first step in the process is to dissolve as completely as possible the nuclear material contained in the irradiated fuels. They are generally enclosed in tight cladding, and the fuel must first be bared by mechanical cutting of the fuel rods, most often accomplished by shearing the assembly.

The actual dissolution is in nitric acid, generally boiling. The uranium, plutonium, and most of the elements contained in the fuel are in this way dissolved, except for:

- Gaseous (Kr, Xe) and volatile (iodine) fission products that evolve in the gaseous effluents from the dissolver. The gaseous flow is treated before discharge into the atmosphere.

- Fission products of the platinum (Ru, Rh, Pd), technetium, and molybdenum family form insoluble intermetallic compounds called dissolution fines (sub-micron particle sizes). In addition, some elements form compounds that are relatively insoluble in a nitric medium. Examples include molybdenum and zirconium, part of which precipitates during dissolution in the form of a deposit consisting basically of zirconium molybdate. These solid particles are separated by conventional centrifugation and filtration techniques; the residue obtained is a few parts per thousand by weight.

The dissolution does not concern the fuel cladding elements and various other structural parts; because of their composition (stainless steels, zircaloy, etc.), they remain unchanged by this operation.

The operations of extraction of uranium and plutonium by tributyl phosphate are at the heart of the PUREX process. In the nitric solution, the elements exist in the following species:

- Uranium is present in degree of oxidation (+VI), in the form of uranyl ions, UO_2^{2+}.
  Plutonium is present basically in degree of oxidation (+IV).

- Most of the minor actinides, together with a very large share of the fission products, are present in degree of oxidation (+III). There are exceptions, such as the alkalis (+I) (Cs, Rb), the alkaline earths (+II) (Sr, Ba), and others to be specified later.

Tributyl phosphate $(C_4H_9O)_3PO$ is relatively immiscible in aqueous solution. Because of the high electron density on the phosphoryl oxygen atom, it has the property of engaging some metallic cations in a coordination complex by an ionic bond. The mechanism is that of solvation:

$$\mathbf{M^n + nA^- + x\ TBP \rightarrow MA_n,xTBP}$$

where M is the metallic element extracted and A- the anion co-extracted, generally the nitrate ion extensively present in the medium.

TBP turns out, generally, to have a much higher affinity for elements in degree of oxidation (+IV) and (+VI) than for trivalent elements.

Thus, the uranyl ion and the plutonium ion will be significantly extracted, while the extraction of the minor actinides and of fission products will be very limited, even negligible in some cases.

Once the elements have been extracted, simply changing certain chemical conditions (reducing the nitrate content of the medium, or reducing the plutonium in state (+III) suffices to reverse the phenomenon and put the species previously extracted back into aqueous solution. The PUREX process accordingly uses successive cycles of extraction and de-extraction of uranium and plutonium to recover and purify these elements.

This description is schematic, because the reality is more complex and more nuanced. Some fission products, such as zirconium (one of the more abundant), are significantly extractible by tributyl phosphate, while others, such as technetium and ruthenium, are present in solution in forms that endow them with unusual behaviour. This is why the extraction/de-extraction cycles must be repeated several times to attain the desired separation factors. The diversity of species present is one source of complexity,

but there is another, just as obvious: the presence throughout these operations of a significant, often even very high, level of radioactivity. The radiosensitivity of some species and of the extractant means that additional separation operations must be performed to isolate the harmful by-products generated and allow recycling of the reagents used. The strict need to limit secondary wastes makes these recyclings one of the conditions of industrial viability of the process.

All of the minor actinides and almost all of the fission products initially present in the spent fuel are grouped in a single "high-activity" solution by combining various flows from the separation process, which are concentrated, and then stored in vessels that are agitated, cooled, and ventilated at all times.

After this storage, which lasts from one to a few years and serves to take advantage of the decay of radionuclides having short half-lives, the high-activity solution is treated to immobilize the wastes it contains and confine them in a matrix that is as stable as possible. In the three countries (France, the United Kingdom, and Japan) that use the PUREX process on an industrial scale, a process of vitrification, in other words of incorporation in a solid matrix having a vitreous structure, is used.

In addition to their long-term confinement qualities (radiation stability, thermal stability, resistance to weathering), vitreous matrices have the advantage of being able to incorporate in their basic structure the great variety of chemical elements present in the processed solution, with an incorporation rate high enough to yield substantial reductions of volume.

The formulation of the glasses must be matched to the chemical composition of the waste; it must produce a matrix that has adequate confinement properties and that can be produced using processes and technologies compatible with the constraints of the nuclear industry.

As a general rule, the glasses used are borosilicates with various additives (Al, Na, etc.) to optimize their properties.

Implementation of the process

The PUREX process is today used on a large scale in France, in the COGEMA complex at La Hague (UP3 and UP2-800° plants having a treatment capacity of 1,600 tonnes of spent fuel per year, values conventionally expressed in mass of irradiated uranium), in the United Kingdom, in the THORP installation (1,000 tonnes/year) operated by BNFL, and soon in Japan, in the Rokkashomura plant (800 tonnes/year).

Attaining these large reprocessing capacities has often required the use of equipment allowing continuous operation. In addition, the constraints imposed by work with highly reactive materials, some of which are also fissile, have led to the development of new equipment.

This has been the case in particular for:

- **The dissolution**, for which the concept of a rotary dissolver including a bucket wheel turning step by step, partially immersed in the solution of boiling nitric acid, was developed. This apparatus simultaneously loads the lengths of fuel to be dissolved, dissolves them, evacuates the resulting solution, and unloads the lengths of empty cladding.

- **The solvent extraction operations:**

The mixer-decanters routinely used in the conventional chemical industry were, historically, the first contactors used in the nuclear industry. While they have definite advantages in terms of effectiveness and stability of operation, they also have serious drawbacks for some operations: they are difficult to adapt to a sub-critical geometry and respond poorly to the presence of solids in the solutions. In addition, the hold times of the liquid phases are very long (several hours), leading to substantial degradation of the solvent.

For operations involving solutions that are highly radioactive or heavily laden with fissile materials likely to induce a risk of criticality, preference is given to the use of pulsed columns, which are better suited to these conditions.

The annular geometry serves to avoid the risks of criticality, the packing with baffles accommodates the presence of solids in the liquid phases, and the hold times of a few minutes considerably reduce the degradation of the solvent even in the presence of very active solutions.

A third type of contactor is also used: the centrifugal extractor. In this type of device, the centrifugal force considerably shortens the decantation times of the emulsions. Their great advantage lies in even shorter hold times than in pulsed columns (a few seconds), making the degradation of the solvent practically negligible. Furthermore, they are extremely compact devices, unlike the other two types of contactor. However, they have the drawback of being revolving devices - they require more maintenance than static devices and are more delicate to adjust. These devices have been chosen for the new R4 plutonium purification shop, expected to be in operation at La Hague by the end of 2001.

- **Vitrification**
  Unlike foreign processes based on large-volume ceramic furnaces heated by electrodes using the Joule effect, the French vitrification process uses a small-volume metallic melting furnace heated by induction. Its limited capacity generally makes it necessary to operate several lines in parallel, but this design has the advantage of greater operating flexibility and allows operation in cells of modest size well suited to the conditions of remote action.

  The resistance of the component materials of the melting furnaces imposes a maximum temperature of 1100-1150°C. This limitation is an obstacle to the development of new glass formulations that might make it possible, for example, to increase the rate of incorporation of radionuclides in the matrix, or more generally to extend the scope of application of vitrification.

  The technology of the direct induction cold crucible should meet these needs for evolution; its principle is induction heating not of the crucible but of the very material to be melted. This makes it possible to keep the wall temperature low enough. Insulated by a thin layer of solidified glass, the wall is perfectly protected from the aggressiveness and temperature of the molten glass.

Operating assessment of the process

The PUREX process, then, achieves the desired separations through a series of operations. The following products are obtained at the end of the reprocessing (figure 1.1 and figure 1.2):

1. Recyclable uranium and plutonium products, with a very high degree of purity. The purification factors attained are of the order of 10^6.
   These products are conditioned in the form appropriate to the recycling specifications (plutonium dioxide, for example, for the production of MOX fuel).

The recovery efficiencies of these two elements are also very high. Values of 99.88% are obtained in the plants at La Hague.
It should be emphasized that this performance is now attained in two steps, rather than the three that hitherto seemed necessary.

2. The minor actinides and almost all of the fission products, conditioned in vitreous matrices as mentioned above. These vitrified wastes group approximately 99% of the $\beta\gamma$ activity of the spent fuel and 99.8% of its α activity (after deduction, of course, of the α activity of the recyclable Uranium and plutonium products). The volume of the vitrified wastes is of the order of $0.1m^3$ per tonne of waste fuel processed.

3. "Structural" wastes (pieces of cladding, mechanical parts of assemblies) that contain the bulk of the neutron activation products formed in the course of the irradiation. They account for about 1% of the $\beta\gamma$ radioactivity.

To these three main products should be added:

- Technological wastes associated with the implementation of the process: most of them exhibit only a low level of radioactivity and are currently stored on the surface.

- Liquid or gaseous flows containing some particular radioelements, such as krypton 85, iodine 129, carbon 14, and tritium. The management mode currently utilized in France for these radioelements is dilution either in the atmosphere or in the marine environment, within the limits permitted by regulations.

- The other elements present at extremely low concentrations in the effluents discharged into the biosphere, within the limits permitted by regulations. The activities discharged are constantly being reduced, and are of the order of 10^{-6} times the radioactivity processed (for the $\beta\gamma$ emitters) and 10^{-8} times the radioactivity processed (for the α emitters) (fig. 2).

Prospects of evolution of the treatment process

Given the technical objectives assigned to the treatment operations, recalled in the introduction, it can reasonably be claimed that the process accomplishes this mission perfectly.

The progress made in recent decades, both in mastery of the chemical mechanisms involved and in the techniques of implementation of the process, has made it possible to achieve the performance stated in the previous section even more safely.

In that case, what changes are needed?

- The first need is for the flexibility needed for the reprocessing of tomorrow's fuels: more highly irradiated fuels, MOX fuels, etc.

- The second need is to continue improving the process, not by further improving its global separation performance, which is already excellent, but by attaining equivalent performance at lower cost and with an even smaller volume of wastes produced and effluents discharged. There are many approaches, ranging from the use of totally destructible reagents to simplifications of the process that would make it possible to increase the unit performance of each elementary operation.

For example, treatment in a single extraction/de-extraction cycle can now be seriously considered.

- The third need could lie in a change in the separation needs themselves, in other words separating elements the PUREX process does not currently separate. It might for example turn out to be worthwhile to separate elements for their recovery value outside the context of the nuclear industry. But it is more particularly the idea of selective management of radioelements having long half-lives that might be realized in this way. This point will of course be taken up again in the part on researches concerning the management of radioactive wastes. It should merely be pointed out here that the PUREX process itself could, with a few adjustments, separate some radioelements having long half-lives, such as iodine, neptunium, and technetium.

Appendix 2: Current status and future R&D program of High-Level Waste Management in France

INTRODUCTION

France has launched a process of optimizing waste management by separating and recycling recoverable energy materials, reducing, conditioning and storing final waste.
Once operations in the back-end of the cycle have been completed, long-lived highly radioactive waste (vitrified waste) represents the major portion of the waste's total radioactivity (99%), conditioned in a small volume (~ 150m^3/year of glass packages).
Nevertheless, the radioactive content of the waste constitutes a potential risk for human beings and environment. The French Law of December 30,1991 set the major orientation for the public policy in the field of HLLW management, and constitutes the first global legislative instrument in that field. It prescribes the research areas to be explored in order to exploit the broadest range of scientific and technical expertise and to provide the elements needed to allow choices on how best to manage long-lived highly radioactive waste. Three areas of R&D were set out by the law :

- minimization of the quantity and toxicity of waste, by studying their partitioning and transmutation;

- packaging and conditioning of long-lived radioactive materials in order to ensure safe and long-lasting containment, and also studying long-term storage;

- evaluation of the feasibility of a deep geological disposal whether reversible or irreversible; underground laboratories are essential tools for this research program.

Public authorities have designated a pilot for each area : the French Atomic Energy Commission (CEA) for partitioning-transmutation, and for conditioning and long term storage, and the National Radioactive Waste Management Agency, Andra, for disposal in geological formations.

The law defined a calendar which stipulates that a comprehensive report evaluating research will be submitted to the French Parliament in 2006, and decide whether it is appropriate to create a repository for HLLW.

Research is conducted in the framework of cooperation among CEA, Andra, partners in the nuclear industry – EDF, Cogema and Framatome – and in research – the French National Council for Scientific Research, CNRS, and universities. It benefits from significant cooperation at the European and international levels. It is constantly evaluated by the National Evaluation Commission, which draws up and publishes an evaluation report annually.

The objective is to identify a set of complementary scientific and technical solutions which serve to define open and flexible strategies for the back-end of the cycle and lay the groundwork for a decision in 2006.

This presentation points out :

1) that very significant results have been produced by R&D since 1991, and that the next steps are clearly defined and well in progress,

2) That, at the present time, we can say that technical solutions for long term management of HLLW do exist, and that the three R&D areas have to be regarded as complementary means, and not in opposition, in a progressive implementation outlook.

MAIN RESULTS ACHIEVED AND OBJECTIVES TARGETED

Minimization of the quantity and toxicity of waste

As a first result, research conducted since 1991 by CEA and Cogema has already made it possible to reduce the quantity of high- and medium-level radioactive solid waste by a factor of three in spent fuel reprocessing at the La Hague facility, and to divide the amount of liquid releases by ten .

Studies concern partitioning (enhanced chemical partitioning during reprocessing), transmutation (transformation in industrial or specialized nuclear reactors into elements which are not radioactive or have a much shorter life), specific conditioning (incorporation of separated elements, which could not be transmuted, within the crystalline network of almost unalterable materials on a time scale characteristic of disappearance through radioactive decay) of the main long-lived radionuclides[1] present in highly radioactive waste.

Concerning partitioning, if the separation of uranium and plutonium and the other fission products depend on a mature chemical process, intensive separation of minor actinides and certain fission products was not possible using industrial processes. Through studies conducted since 1991, a reference process scheme was defined for an advanced partitioning process for the main long-lived radionuclides :

- neptunium and iodine could be separated by adapting the PUREX process used industrially in the reprocessing facilities at La Hague,
- To separate americium and curium, it was necessary to develop new chemical separation processes by developing very selective molecules capable of separating these elements.

The families of extractors were defined, the principal reference molecules synthesized, and their performances verified experimentally (partitioning ≥ 99%) on real radioactive solutions in order to reach the stage of scientific feasibility (2001).

The next stage will be that of technical feasibility, moving from the molecule to the overall chemical process, which will be defined and validated in 2005. The existence of the reprocessing industry makes the implementation of these processes a real possibility.

With reference to transmutation, research conducted in recent years has pointed up the performances of transmutation in different types of electronuclear power plants, demonstrating the capability of multirecycling the plutonium in PWRS, with improved plutonium fuel assemblies, allowing to consume all the plutonium which is both an energetic material and the main contributor to long term radiotoxicity. Results have also pointed and that by separating and multirecycling the actinides in a reactor, the mass and toxicity of the waste, at equilibrium, are divided by 100 compared with the equivalent in an open cycle, and that innovative reactors (electricity-generating reactors or those dedicated to transmutation) would present characteristics adapted to these performances (great capacity to consume plutonium as well as

[1] Minor actinides and certain very long-lived fission products abundant in spent fuel and potentially more mobile in the environment.

long-lived radionuclides and ability to use the best advantage the energy contained in the fuel: rapid spectrum, fuel with a very high combustion rate, reactor-fuel integrated cycle,…).

In this field, let us note that experimental studies on fuel for transmutation in fast neutron reactors particularly comprise an irradiation program in the Phenix reactor, which is at present under inspection, renovation and maintenance, in view of restarting at power operations in 2002 for six irradiation cycles.

Conditioning-Storage

Research is conducted in two areas :

- the development and acquisition of knowledge on packages (conditioning, long-term behavior and characterization),
- The definition of concepts for interim storage facilities, above or under ground, their evaluation and related research and development.

In particular, developments have made it possible :

- To establish the scientific basis for a true science of the long-term behavior of packages in which, particularly for vitrified waste packages, improvements in scientific understanding and the description of phenomena made it possible to produce a long-term evolution model that indicates an alteration of the glass of approximately one ten-millionth per year (i.e 99,9% of the glass would be intact after 10,000 years of alteration in the event of interaction with water),

- To develop concept for multi-use containers for dry long-term storage of spent fuel allowing their reopening in safe conditions for later reprocessing or geological disposal . Special attention is paid to the functions of leaktightness, inspection, resistance to hazards, loading and closing operations. Functions which require testing will be examined in 2002, on appropriate test benches, and full-scale objects will be produced in 2004 to qualify all of the fonctionalities.

Research on long-term interim storage, above or under ground, mainly focuses on spent fuel that would not be reprocessed in the short term, and packages of high- and medium-level waste. This work is based on an analysis of the essential functions of long-term interim storage (durability and safety of containment over a long time period while preserving the integrity of the packages, recovery under safe and technically established conditions). This leads both to the definition of efficient concepts and to the scientific and technological demonstration of this performance, and to the proof of the capacity of interim storage to fulfill its functions for a seculary time scale.

Two main families of concepts have been developed, above and under ground, and they comprise specific modules respectively adapted to the storage of spent fuel, high-level radioactive waste and medium-level radioactive waste .

Research is conducted to support the scientific demonstration in order to develop the knowledge required for the demonstration of performances in terms of the strength and durability of the interim storage facilities : study of the long-term behavior of infrastructure materials (concrete, site materials), study of the influence of site effects on the seismic hazard of interim storage above or under ground.

Deep geological disposal

The French Government has indicated in 1998 that it was in favor of the option of reversibility, and published in 1999 the official authorization for implementing the underground laboratory at the boundaries of the Meuse and Haute-Marne departments, in the east of France, in a sedimentary clay geological formation, 150 million years old, 130 m thick, and about 500 m deep. This formation shows very good geological continuity and stability, low permeability and suitability for mining drilling techniques.

A key achievement was the beginning of the first excavation work for the laboratory in August 2000. Shaft sinking progresses according to schedule reaching a depth of ~ 40 m at the beginning of 2001, the drilling equipment required to reach the main level of the laboratory at a depth of 490 m underground are currently being set in place .

During year 2000, Andra also studied the underground laboratory expansion, using 3D seismics, as well as borehole observations, and measurements have confirmed the interest of the considered clay formation for futher investigations.

At the same time, a significant effort was made to organize the necessary models and simulations for the study of geological disposal feasibility, by identifying and prioritizing the various phenomena to be taken into account in the typical repository situations, and by implementing the necessary environment for code integration and coupling.

Concerning the scientific and technical program in the underground laboratory, it has four main objectives : knowledge of the geological medium, characterization of the geomechanical properties of the clay formation, assessment of its confinement properties, and long term evolution of these properties. Andra aims to produce these results in a report to the Government in 2005.

Besides this progress at Bure, on the other hand, due to local oppositions, the Government suspended in June 2000 the work of the administrative “Granite Mission”, which had been mandated for identifying possibilities for siting a second underground laboratory in granite formation.

As result, the Government stated in July 2000 that decisions could only be reached on HLLW management if all necessary research could be carried out.

CONCLUSIONS AND OUTLOOK

The goal of this research is to propose waste management options which will inform the decisions of the French Parliament and Government in 2006, based on the following main guidelines : minimization of waste, confinement and reversibility.

Solutions do exist, that could be implemented in a progressive manner and several scenarios might be considered, for instance :

- plutonium consumption first, because it is the major contributor to the radiotoxical inventory and therefore its final disposal might be avoided ; the development of advanced plutonium fuels (CORAIL, APA) would make it possible to recycle all the plutonium in PWRS , the final waste (FP, MA) being vitrified, ensuring a long lasting confinement,

- a possible step further could be to implement the partitioning of the minor actinides (which present the highest level of long term radiotoxicity after plutonium), as an extension of the existing reprocessing capability; the result would be that the new vitrified packages would contain only mostly fission

products, and would this come down to the radiotoxicity level of natural uranium from the mine in some hundred years; the minor actinides could be conditioned in a form that would allow for later transmutation, while very efficient containment is ensured,

- concerning transmutation, future nuclear power production systems are being studied incorporating the objective of waste minimization, with the design and evaluation of new systems (reactor and fuel cycle) and the development of the related key technologies,

- in any case, a geological repository might be useful when regarding long term burden free solutions, to dispose the ultimate wastes, benefiting from the significant reduction in radiotoxicity provided by reprocessing, recycling and transmutation; the France R&D program in this field is in good progress;

- The law of December 1991 has also required to consider the long-term storage, in order to have several open options and bring flexibility in the implementation with time of the solutions for the long-term management of radioactive waste.

Thus, and this is mandatory, several solutions will be available for presentation to the Government and the Parliament, for open debate and choices of options for the long-term management of HLLW in France.